# Fully R2R-Processed Flexible OLEDs for Lighting

***Takashi Minakata, Mitsuru Tanamura, Yasuhiro Mitamura, Masayuki Imashiro, Akira Horiguchi, Akira Sugimoto, Masahiko Yamashita, Kento Ujiiye, Satoshi Sunahiro, Yukito Yada, Nobuki Ibaraki, Hiroshi Tomiyasu***

**CEREBA, Tsukuba, Ibaraki, Japan**

## Abstract

*Flexible organic light-emitting diodes (OLEDs) on continuous plastic film have been successfully fabricated using full roll-to-roll (R2R) processes, including depositions of gas-barrier layers, electrodes, layered-organic semiconductors, and lamination encapsulation. The OLEDs exhibit excellent stability and high performance comparable to OLEDs on glass, with the incorporation of R2R deposited high-gas-barrier layer with a WVTR below $10^{-5}$ g/m$^2$day. Also a strong correlation between the gas-barrier performance and the stability of the OLEDs stored in high humidity environment has been confirmed.*

## Author Keywords

Flexible OLED; roll to roll (R2R) fabrication; gas-barrier film; WVTR; dark spot growth.

## 1. Objective and background

Recently, flexible OLEDs have attracted significant attention as regards emerging applications in both lighting and displays. The roll-to-roll (R2R) process is considered to be the ultimate solution for flexible device fabrication for large scale production, since traditional processed OLEDs based on the sheet-to-sheet (S2S) process have become almost saturated. In addition, there are advantages to R2R system as regards product transport and storage. Technical trends for wearable smart devices have added to accelerated demand for flexible devices, and encouraged the development of enlarged new device applications, and broadened the market for newly designed or unusual and unique lighting applications using flexible OLEDs. Active R&D projects on R2R-processed OLEDs have been reported by several European research institutes [1-3].Now the commercialization of flexible OLEDs by the R2R process expected and will be realized shortly. These developments have therefore motivated us and to fabricated fully R2R-processed OLEDs for lighting for the first time using R2R techniques from pre-treatment, barrier formation, core-layer deposition to encapsulation.

Printed OLEDs are expected to be used in future device fabrication to realize rapid fabrication and reduced material consumption, however fully printed OLEDs have not been established to date. This is because the performance of these devices is inadequate compared with that of dry-processed OLEDs. A realistic solution, balancing both performance and processability is assumed to combine both wet and dry processes in R2R fabrication. Attempts to introduce both fabrication lines have been made and compatibility issues have been examined.

Gas-barrier technologies both fabrication and evaluation are indispensable for the development of flexible OLEDs formed on plastic film substrates. A reduction of several orders reduction of magnitude in the permeation of plastic film substrate, specifically $10^{-6}$ g/m$^2$day of water vapor transmission rate (WVTR), is required to protect the OLEDs and to extend the life for several years. The detection of ultra-high gas barrier has almost been established [4], however, the correlation between high gas barrier performance and real stability of OLEDs has not been determined. IN this paper, various kinds of barrier layers ranging from WVTR values of $10^{-3}$ to $10^{-6}$ g/m$^2$ day are fabricated, and the stabilities of the OLEDs on the films with these barriers are studied.

## 2. Fabrication procedure

### (a) R2R fabrication lines

R2R fabrication lines of this study consist with pre-treatment, gas-barrier layer formation, vacuum deposition and encapsulation as presented in Figure 1. All the fabrication lines are R2R-style and are installed in a clean room of class 10,000. Therefore, the handling and transfer of fabrication components such as substrate rolls, evaporation sources, and sputtering targets were conducted under a controlled clean environment.

### (b) Pre-treatment

In order to eliminate or reduce absorbed particles and/or impurities on the film substrate surface, the rolled film was subjected to pure-water washing and/or UV irradiation, since ordinary widely available plastic film roll contains particles that form defects on the OLEDs. Furthermore, in order to obtain a smooth surface, a smoothing layer was deposited using a wet-processed slit-die R2R coater. The slit-die coater can provide precise part-time coating with a highly controlled coating solution volume flow. Although the drying speed is limited in our system, it is possible to accelerate the operation speed to modify the drying capability. For the coating of the OLED layers, very thin layer of below 100nm in thickness must be deposited, and at the same time, thickness uniformity in the lighting area must be maintained in order to achieve uniform light emission. This wet coating apparatus can control the wet-coating process precisely and it used for the wet-coating of semiconductors For example it is applied in the fabrication of hole injection and light-emission materials, and attempts to form wet-coated layers are in-progress.

### (c) R2R gas-barrier-layer formation

It is well understood that common plastic film allows moisture to penetrate very rapidly, which will degrade OLEDs, so formation of a gas barrier layer on plastic film substrate is required to protect the OLEDs. Two kinds of barrier-layer deposition methods, plasma enhanced chemical vapor deposition (PE-CVD) and atomic layer deposition (ALD) were used in our system. PE-CVD is effective for the deposition of barrier layer with a high deposition rate and ALD is effective to compensate defects on film surface because of its high coverage. Both barrier layer depositions are performed at ambient temperature or slightly raised temperature of below 100 ℃.

### (d) R2R vacuum deposition

The core stages in the OLEDs fabrication are the vacuum-processed steps described below. Each step is aligned and connected under reduced pressure. In this study, the rolled film

was first installed in unwinding unit, and the installed chamber was evacuated. It is important that degassing of the plastic film is performed in order to obtain a high-vacuum environment and to avoid degradation of OLEDs. Note that in our system, heating component has been installed in the vacuum line to release gasses effectively. After out-gassing, anode electrode was deposited using sputtering. Subsequent depositions of organic semiconductor layers namely the hole-injection transport, light-emitting, electron-transport layers were then performed. Finally, the cathode electrode was vacuum-deposited. In some cases, the thin film encapsulation of silicon nitride by sputtering was applied to the cathode-deposited surface for passivation. These deposition processes were performed under high vacuum apart from the sputtering.

### (e) R2R encapsulation

Film lamination was then applied to the fabricated OLEDs, performed under ambient pressure. In order to transfer the films from the vacuum zone, a pressure-controlled unit was required to connect both the vacuum and ambient pressure units. A pressure- controllable chamber was installed between two units to collect the OLEDs layer-deposited films, and connected with gate valves at each ends. Cap-film lamination was then conducted using roll-lamination with heating under a controlled nitrogen atmosphere, along with partial evacuation to eliminate bubbles from the contact region. After the cap-film lamination, the OLED roll was wound and removed from the encapsulation line. Flexible OLEDs have been fabricated with four individual R2R lines, washing (pre-treatment), wet coating, barrier-layer formation and OLEDs layers formation & encapsulation.

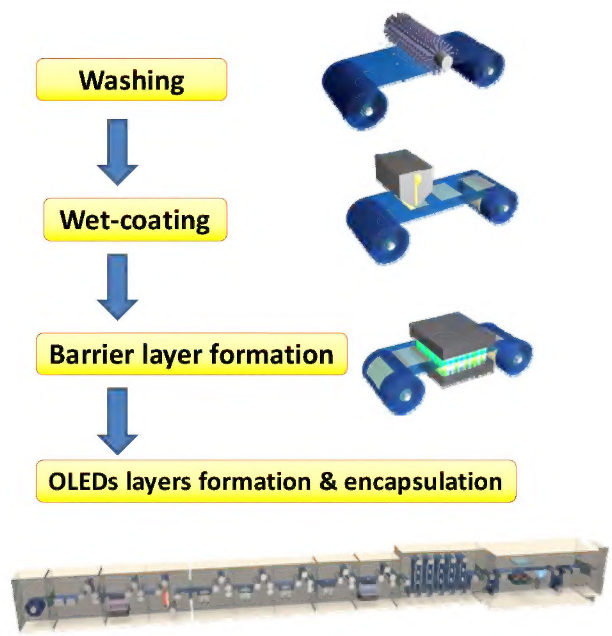


**Figure 1.** R2R fabrication lines

## 3. Flexible OLED fabrication and performance

OLEDs were fabricated on polyethylene naphthalate (PEN) substrates with a thickness of 125 μm, width of 300 mm, and film length above 150m. Firstly, the PEN film was washed in order to remove dust particles from the film surface. Several types of washing processes were applied, including water blushing, sonication in pure water, nozzle-spraying, and exposure to UV light followed by drying. The cleaned film was rolled and transferred to a wet-coating line. A planalized polymer layer was coated using the slit-die method to form a smooth surface on the substrate, which was assumed to be effective in reducing defects on both the barrier layer and the OLEDs. Subsequently, a SiOC barrier layer with thickness of approximately 300 nm was deposited on the R2R-designed PE-CVD using hexamethyldisiloxane (HMDSO) as a source material. In addition, an alumina layer with a thickness of 20nm was deposited using R2R ALD to prevent pin-holes appearing in the barrier layer.

The entire OLED layer fabrication process was conducted in vacuum. The barrier-layer-coated film was installed in the R2R vacuum chamber and out-gassing was performed by passing the substrate through the heating zone to remove absorbed moisture and gasses in the film. The level of out-gases and the gas species from the film was monitored using Q-Mass spectroscopy. An anode electrode of indium zinc oxide was deposited using sputtering with a thickness of 150 nm. Then the OLED layers of HIL/HTL/EL/ETL/EIL were formed with stepwise depositions and finally coated with Al with a thickness of 100 nm as a cathode electrode. In the OLED structure, green phosphorescent material doped in the host was applied to the EL layer.

After the formation of OLED layers, the film was forwarded to the accumulation chamber and venting with N2 to atmospheric pressure. The film was transferred to the encapsulation chamber and laminated with PEN/Al as a cap film. The encapsulation chamber was under a controlled N2 atmosphere to avoid degradation of the OLEDs. As previously stated, entire process from evacuation to encapsulation was R2R-connected.

A photograph of the OLEDs fabricated using the R2R process is shown in Figure 2. Each light emitting area of 30 mm square is aligned with the continuous film substrate. The typical current efficiency was approximately 30 cd/A, which was comparable to or larger than that of conventional OLEDs formed on glass substrate. This indicated that the reduction of performance of the R2R-fabricated flexible OLEDs was negligible. In addition, a slight enhancement of current efficiency was assumed to be an improvement of light extraction, due to the out-coupling structure in the film OLEDs.

**Figure2.** Photograph of R2R processed OLEDs lighting area

## 4. Fabrication complexities

Manufacturing complexities in the R2R process are a major concern as regards the mass production of flexible OLEDs. Film

transfer was one of the most prominent issues, which was assumed to be caused by electrostatic charging, film deformation due to tension applied to the film, and/or heating during the process. Electrostatic charging during film handling occurred even during contact with the metallic roll, which was observed in form of film wrinkling or light sparking by discharging. The electrostatic charging was evaluated by installing a contactless measurement unit into the chamber, and .electrostatic charging up to several kV on the film was confirmed, which depending on the film position. The degree of the charging is assumed to be influenced by the film material and surface conditions, which should be controlled in the manufacturing lines. Another important issue is damages to the OLEDs during the fabrication process. Assumed damage factors are moisture incorporation, high temperature heating, mechanical stress on film transferring, etc. The influence of each factor is assumed to be dependent on both OLED materials and device structures. Therefore, it is necessary to develop highly stable materials and to design stable structures against process damage.

## 5. Performance difference between S2S and R2R

The conventional fabrication of flexible OLED devices is conducted using the sheet to sheet (S2S) process. In order to evaluate and compare the R2R and S2S process characteristics, flexible OLEDs were fabricated using the S2S process. The resultant device performance was compared with that of the R2R processed devices as listed in Table 1. In the early R&D stage, R2R-processed devices showed poorer performance than those of S2S-processed devices. Then we examined the influence of various fabrication process conditions, and thus optimized for the R2R fabrication conditions. As a result, the R2R devices exhibited the same performance with the S2S devices. The details of the tuning process will be presented at the conference.

**Table 1.** Relative light emission efficiencies of flexible OLEDs to rigid OLEDs on glass

| Device | Current efficiency | Quantum efficiency |
|---|---|---|
| R2R | 105 | 108 |
| S2S | 100 | 100 |
| Rigid/ glass | 96 | 99 |

## 6. Gas-barrier performance and storage stability

In this section, two important issues of barrier-layer performance to protect moisture penetration and mechanical stressing on the flexible devices are examined. In order to evaluate barrier-layer performance, several kinds of OLEDs with different barrier-layer structures were fabricated and WVTR measurements on each barrier layered film were made.

The layered barrier films were formed in the batches combined with PE-CVD deposited silicon oxide, sputter deposited silicon nitride, and ALD deposited aluminum oxide, and a selection of deposition conditions. The barrier-layered films showed high barrier performance, ranging from $10^{-3}$ to $10^{-6}$ g/m$^2$day of WVTR at 40℃ and 90% relative humidity, as measured by well-established measurement apparatus system of ambient pressure mass-spectroscopy(API-MS) and cavity ring-down infrared spectroscopy(CRDS).

The OLEDs formed on the various barrier coated films were stored in 65℃ and 90% relative humidity and dark area growth in light emission surface was observed using optical microscopy. Photographs of OLEDs lighting surfaces in 2 mm square observed using the microscopy after storage are shown in Fig 3. The enlargement of dark areas on the OLEDs with poor barrier layer is distinct, while the growth of dark area on the highly barrier-layered films is negligible. The dark area ratio of the OLEDs was plotted against the stored times and the various barrier-layered OLEDs ware compared as shown in Figure 4.

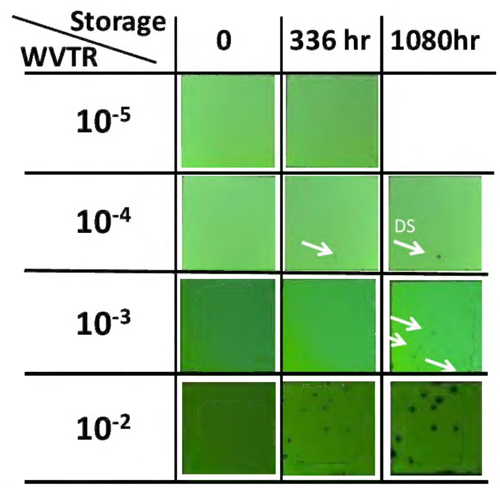


**Figure 3** Changes in light emission areas of OLEDs upon storage time in 65°C, 90%RH.

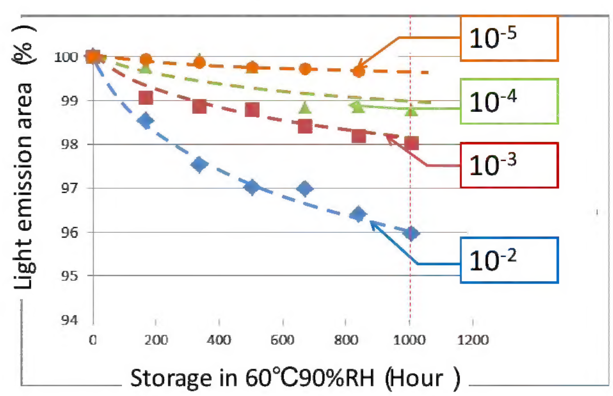


**Figure 4** Dark area ratios to storage time of OLEDs with different WVTR

A strong correlation between barrier performance and dark area growth is confirmed. Storage for 500 hours in 65℃ with 90% relative humidity corresponds to 20,000 to 50,000 h storage in ambient storage at 30℃ with 50% humidity. This indicates that a barrier performance below $10^{-5}$ g/m$^2$day is sufficient for practical OLED devices. Penetration routes from the sealed edge was possible, however this behavior was assumed to have minimal influence on dark area growth, since the length of the

seal edge was sufficiently large enough in the OLEDs. Further elucidation of the gas-penetration mechanism on encapsulated flexible OLEDs is subject for extensive research and an important issue for flexible device design.

## 6. OLED flexibility

Of the several advantages of flexible OLEDs, flexibility is the most important characteristic as regards film-shaped OLEDs. The authors have tested mechanical stress reliability for flexible OLEDs. Several kinds of mechanical stress were had evaluated since these forms of stress are applied during R2R processing, storage, transport, and for emerging applications. Typical mechanical stress modes include bending, rolling, twisting, and extraction and schematic images for these types of mechanical stress are shown in Figure 5. Bending is most fundamental mechanical stress experienced in flexible OLEDs, and bending conditions such as the bending radius, bending speed, duration time, and degree of repetition are essential parameters for mechanical stress testing. A photograph of twisting stress test apparatus is shown in Figure 6. For lighting applications, mechanical stress conditions are relatively moderate compared with those of display application, since even in rolled or bend lighting bending radius is assumed to be of the order of several cm. During testing our flexible OLEDs, periodic bending was conducted for 100 times and degradation was not observed. In addition, mechanical stress tests with rolling and tension reflecting from R2R fabrication conditions were conducted and no degradation after the stressing was observed.

These results indicate that the degradation of the OLED devices during R2R fabrication process is negligible. It is assumed that the bendability of flexible device can be tuned by composed materials, since the volume fraction of the thin film OLED layers is very small compared with those of substrate and encapsulation materials. Fabrication processes involving thin substrates and/or thin encapsulation materials and evaluation of mechanical stress durability of such devices are in progress. In contrast, from the perspective of the R2R fabrication process, a very thin film substrate could lead to difficulties such as elongation with tension, wrinkle formation during transfer, etc. Therefore, the flexible OLED device structure must achieve balance between device application and fabrication feasibility of the R2R process.

## 7. Conclusion

Flexible OLEDs were fabricated using the fully R2R process including barrier layer formation, core layer deposition and encapsulation for the first time. The resultant OLEDs exhibited excellent performance with a current efficiency of 30cd/A, which was comparable to OLEDs on glass. The flexible OLED formed with a R2R-deposited high-gas-barrier layer with a WVTR below $10^{-5}$ $g/m^2day$ exhibited stable performance and the correlation between barrier performance and the stability of the devices was confirmed. Mechanical stress testing of the flexible OLEDs was conducted and no degradation due to damage reproducing from R2R process was confirmed.

## 8. Impact of research

We have demonstrated a full R2R fabrication process for flexible OLEDs for the first time, i.e., R2R procedures are used in the pre-treatment, barrier formation, core-layer deposition, and encapsulation stages. The flexible OLEDs show a current efficiency of approximately 30 cd/A, which is comparable to or larger than that of conventional OLEDs formed on glass substrate. Mechanical stress testing is conducted, utilizing stresses commonly experienced by OLEDs during fabrication, storage, etc. It is found that the degradation of the OLEDs during R2R-fabrication is negligible. Most notably, a R2R-deposited high gas-barrier layer is found to yield improved moisture resistance, and a correlation between barrier performance and device stability is confirmed. The findings of this paper are relevant to the OLED fabrication industry, along with the various fields in which OLEDS are applied.

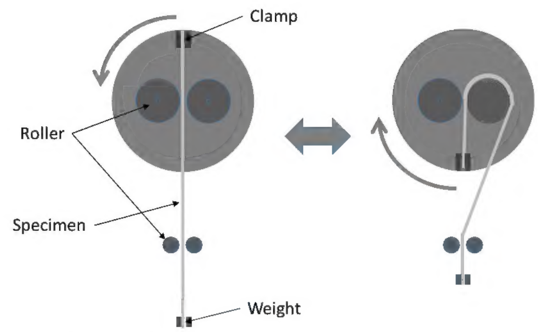


**Figure 5** Apparatus for bending stress testing

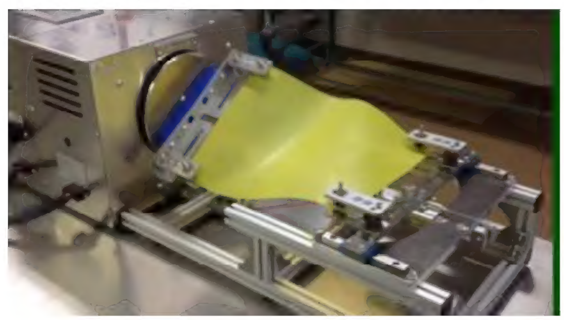

**Figure 6** Photograph of apparatus for torsion testing

## 9. Acknowledgements

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